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ABSORPTION OF MOISTURE BY GELATIN IN A SATURATED ATMOSPHERE[1]

CHARLES A. SHULL AND S. P. SHULL

A great deal of work has been done to determine the relation of colloidal matter to water, gelatin being a favorite substance for such studies. However, one finds but few attempts to determine the water relations established when the colloidal material is exposed to an atmosphere saturated with water vapor. The earliest observations of a difference in behavior of colloids toward water and water vapor are credited to Volbehr (5). The main work dealing with this subject is a paper by von Schröder (3), who claims that gelatin absorbs much more water when placed in liquid water than when exposed to a vapor-saturated atmosphere. The data presented in his paper were afterwards used as a basis for a theoretical discussion by Bancroft (1), who attempted an explanation of the observed phenomena, but without any apparent attempt to verify von Schröder's results. One finds in the recent literature occasional reference to these papers, as in Czapek (2). Here Czapek (p. 42) adopts Bancroft's explanation of the supposed difference between the vapor pressure of the colloid and that of the mass of water which saturates the atmosphere about the colloid.

For the sake of clearness it will be advantageous to state briefly the results of von Schröder's investigations as given in section VII of his paper, which is entitled "Ein Beitrag zur Thermodynamik der Quellung" (*l.c.*, pp. 109–117).

In the first place, gelatin absorbs water very rapidly from liquid water. A piece of gelatin weighing 0.801 g., and which contained 17.6 percent of hygroscopic water, took up moisture as shown in table 1.

TABLE 1. *Absorption of Water by Gelatin from Liquid Water*

Time	Intake in Grams	Gain Percent*
5 mins.	2.282	336.1
10 mins.	2.934	432.1
20 mins.	3.669	540.3
30 mins.	4.072	599.7
40 mins.	4.300	633.3
50 mins.	4.415	650.2
60 mins.	4.506	663.6
2 hrs.	4.941	727.7
24 hrs.	6.911	1018.
48 hrs.	7.734	1039.

* Calculated on absolute dry weight of gelatin disc, 0.679 g.

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[1] Contributions from the Botanical Laboratories of the University of Kentucky, No. 3.

But if a similar piece of gelatin is placed in a saturated atmosphere, moisture equilibrium between gelatin and water is reached at a much lower percentage of intake, as shown in table 2. In this case the air-dry gelatin disc weighed 0.904 g.

TABLE 2. *Absorption of Water by Gelatin in a Saturated Atmosphere*

Time	Intake in Grams	Gain Percent
1 day	0.154 g.	17.08
2 days	0.218	24.10
3 days	0.277	30.69
4 days	0.294	32.56
5 days	0.347	36.21
7 days	0.357	39.52
8 days	0.366	40.52
15 days	0.372	41.18
17 days	0.368	40.74
18 days	0.369	40.80
20 days	0.374	41.41

These figures make it appear that equilibrium was reached at about the end of a week, at a little over 40 percent of absorption.

When the gelatin was first soaked in water till nearly saturated, and then brought into a saturated atmosphere, there was continuous loss of water from the gelatin. Thus a piece of gelatin weighing 0.433 g. was soaked until it weighed 5.092 g. It was then placed in a chamber with a supposedly saturated atmosphere. The behavior is shown in table 3. The first three columns to the left, except the top line, are taken from von Schröder, and the two columns to the right are from Bancroft's discussion of the same experiment, with a correction made by omitting the dashes in Bancroft's table, and lifting the figures into correct alignment with the time intervals.

TABLE 3. *Loss of Water by Gelatin in a Saturated Atmosphere*

Time	Loss in Weight in G.	Loss Percent	Weight of Water in Gelatin in G.	Percentage Absorbed Water Remaining
0 day	——	——	4.659	1,076
1 day	0.259	6.29	4.400	1,016
2 days	0.337	7.82	4.332	998
3 days	0.383	8.87	4.276	988
4 days	0.418	9.68	4.241	979
5 days	0.929	21.50	3.730	861
7 days	1.313	30.45	3.346	759
9 days	1.972	45.63	2.687	621
11 days	2.571	59.48	2.088	482
14 days	3.175	73.46	1.484	343

The percentages in column three are not calculated with great accuracy, but are presented exactly as in the original. The data indicate that there is continuous loss of water from the gelatin, although the air on all sides is assumed to be saturated. The behavior implies a contradiction to the second law of thermodynamics. Such an evaporation of water from one part of the system to another under the conditions could take place only if the colloidal matter had a higher vapor pressure than the liquid water; and as the water evaporated from the colloidal mass, it would have to condense at the surface of the liquid water. Bancroft's explanation of the higher vapor pressure is based upon the shape and size of the water droplets in the gelatin. They are assumed to be round, and it is held that water evaporates more readily from a curved surface than from a flat one. The droplets of water are so minute that the curvature of the surface of each droplet is quite sharp; this results in a vapor pressure higher in the colloid, and a consequent distillation of water from it to the liquid phase of the system. The distillation should go on until in some way or other the vapor pressure throughout the system reaches equilibrium.

Being engaged in the study of the relation of certain colloidal organic substances to water, we have had occasion to perform some experiments which led to a repetition of some of von Schröder's work. However, it was not possible to tell from von Schröder's discussion just how he set up his experiments, how he controlled the temperature, and how he secured and maintained saturation. In a number of ways the discussion leaves one in the dark, and we were compelled therefore to work out a method of investigation which no doubt differs in a number of ways from von Schröder's. The methods employed are briefly stated.

Materials and Methods

The gelatin used in the experiments to be recorded here was the Gold Label gelatin commonly handled by dealers. No attempts were made to purify it in any way; for, although pure substances are to be preferred in original work of any kind, we felt justified in using the gelatin in its commercial condition because the experiments we were trying to repeat had been carried on with unpurified gelatin. There is no doubt that von Schröder's gelatin as well as ours contained acids, and that in both cases salts were present. We could have neutralized our gelatin with some inorganic base, with subsequent dialysis until it was salt-free. This would have given us a purer gelatin, but we could not then have made a direct comparison between our own and von Schröder's results. It seemed to us best therefore to repeat the experiments with ordinary gelatin, as the differences between the two substances would likely be less than if we used any purification method to remove acids and salts.

During the earlier tests the gelatin was prepared by dissolving it at gentle heat in distilled water, after which it was poured into petri dishes which had been slightly smeared with glycerol to prevent the gelatin from sticking to the glass; but as the glycerol is hygroscopic, this method was abandoned to avoid contamination of the gelatin with glycerol. Mercury was used as an agent to prevent the gelatin from sticking to the glass during drying. In this way sheets of gelatin of desired thickness could be secured, and these sheets were cut into disks before they became entirely dry, while still slightly pliable, and the disks were dried out in contact with air until weight loss ceased. During this preparation the gelatin was carefully protected from dust to reduce chances of mold or bacterial infection.

The disks were exposed to a saturated atmosphere in small wide-mouthed bottles. The bottles were arranged with a layer of mercury in the bottom sufficient to sink them. Over the mercury was a layer of water. The gelatin disks were suspended over the water, just as near the water surface as practicable, in shallow paper baskets which were attached by threads and wax to the center of the rubber cork which closed the bottle. The rubber cork was shellacked upon its inner and outer surfaces to protect it from water, probably an unnecessary precaution.

To control the temperature the bottles were sunk in a Freas thermostat which was set to run at 26° C., and which showed no deviation with an ordinary chemical thermometer during the five months while the tests were run. It was found necessary to control the growth of bacteria and molds on the gelatin, and this was accomplished by the use of small pieces of thymol in the water. The bottles were usually placed in the thermostat for some time before the gelatin was introduced. This allowed the air to become more nearly saturated. Then the weighed disks were put into the baskets. At intervals which were purposely made infrequent so as not to interfere with the saturation of the air, the disks were removed carefully and quickly to weighing bottles and weighed. The greatest care was taken to keep the disks from drying out during weighing, and to keep the air to which they were exposed during intake intervals from becoming unsaturated. The bottles were always corked and returned to the thermostat during weighings. It was noted that there was always condensation of vapor on the walls of the weighing bottles, so that it was not possible to prevent all losses of water.

Results

In all cases it was found that much more water was absorbed by the gelatin from the atmosphere than von Schröder had observed. During the preliminary tests in one instance there was a regular intake of water which continued for weeks, and which had reached an intake of 250 percent of the weight of gelatin when it was accidentally overturned by the laboratory attendant, and the experiment was thus brought to an abrupt end. A considerable number of disks were started, some with and some without thymol. The rate of intake was apparently about the same during the first several days, but the disks exposed without thymol would always suffer in time with bacterial or mold infections. Usually after about three days the uncontrolled disks had to be discarded. The indications were that the thymol itself was not noticeably accelerating the rate of water intake. Plotted curves practically coincided during the first several days, with and without control.

One typical example of the intake has been chosen to illustrate the behavior of the gelatin under the given conditions. The gelatin disks weighed 0.787 g. air-dry. The absorption data obtained during 47 days are given in table 4.

TABLE 4. *Absorption of Water by Gelatin in a Saturated Atmosphere.*

Time	Intake in Water	Gain Percent
6.5 hrs.	0.1292 g.	15.27
16.5 hrs.	0.2174	27.62
1.0 day	0.2698	34.28
2.0 days	0.3796	48.23
3.0 days	0.4599	58.43
4.0 days	0.5332	67.75
5.04 days	0.5884	74.76
6.0 days	0.6260	79.54
8.0 days	0.7109	90.20
10.0 days	0.7754	98.52
12.0 days	0.8356	106.17
14.0 days	0.8734	110.97
18.0 days	0.9644	122.54
22.0 days	1.0374	131.81
26.0 days	1.0979	139.50
30.0 days	1.1602	147.42
35.0 days	1.2209	155.13
40.0 days	1.2886	163.73
47.0 days	1.3426	170.59

After the last weighing recorded in table 4 was made, the disks were returned to the bottles and left undisturbed for two months, as they were hard to handle without breaking. On opening them at the close of two months, the disks were found in liquefied condition. No culturing was attempted to determine whether liquefying bacteria might have been present. But the thymol had inhibited development of molds, and it is usually considered that molds are less readily controlled than bacteria. It does not appear to us likely, therefore, that liquefaction was brought about by bacterial action.

The contrast in behavior of gelatin as we have found it, and that reported by von Schröder, is shown graphically in figure 1. The possible cause of this difference will be considered later.

One striking thing which has been noted in regard to water absorption is the regularity of intake. It usually begins rapidly, and becomes continually less rapid as absorption increases, falling off in regular fashion till approaching saturation causes a more rapid decline in the rate of intake. The rate of absorption has been studied particularly with reference to seed colloids soaking in water, and mathematical analysis has given us a formula by which the intake can be closely approximated by calculation.

The same kind of analysis was made of the gelatin absorption from saturated air, and the same formula that was derived from the absorption of water by seeds from liquid water can be used in calculating the curve of absorption of water by gelatin from a saturated atmosphere.

The generalized formula is $y = a \log_{10} (bx + 1) + c$, in which y is the percentage of total intake, and x the time, with a, b, and c constants.

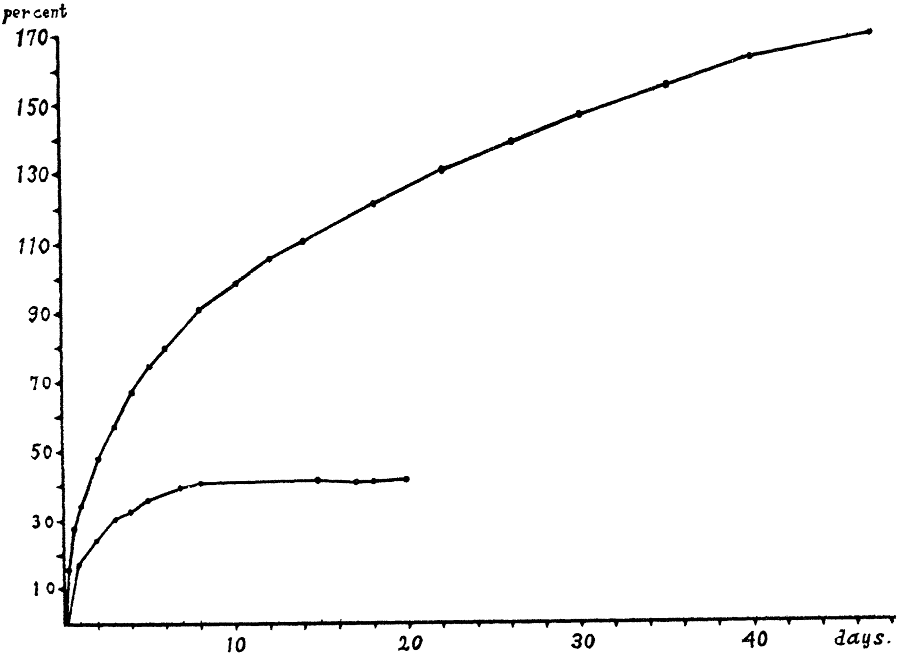

FIG. 1. Curve of moisture intake of gelatin in a saturated atmosphere. Lower curve, from von Schröder's data. Upper curve, from data presented in Table 4.

In the case of seeds we found it necessary to use two or even three curves to approximate the experimental data (4). Similarly it is necessary to use two equations for curves joining in a common tangent value to express the gelatin absorption curve.

The first part of the curve, with the values of a, b, and c substituted in the equation, is as follows: $y = 93.4 \log_{10} (0.032x + 1) + 10.413$; while the later part is expressed thus: $y = 141.9 \log_{10} (0.0064x + 1) + 40.82$. These two curves have equal tangents at $x = 209.47$ hrs., at which time the two values for y are, $y_1 = 93.22723$, and $y_2 = 93.22764$, showing a break in the curve of only .00041 per cent. In other words, at the end of about six days, when the gelatin has taken in something less than its own weight of water (93 percent) it requires different values for the constants in the general formula to keep a calculated curve running close to the data. With these two successive curves there is fairly close agreement between the calculated and the observed data as shown in table 5.

The agreement is very good with the exception of the first and the next to the last readings. The data at 960 hours may be in error, although there is nothing in the records to indicate it. It seems scarcely likely that the data which had been running so regularly during the preceding 500 hours would suddenly rise above the curve, and then drop back again to the curve 168 hours later. If the error involved accidental addition of the water to the gelatin, it should show in the last reading also. If an error was made it was most likely an error in counting the weights, as a reduction of 10 milligrams in the weight would bring the data to 162.46, which would bring fair agreement with the calculated value. But even as the figures stand the agreement is very striking, and shows that water intake goes steadily forward at a rate determined by the conditions of the experiment.

TABLE 5. *Agreement of Calculated and Observed Intake by Gelatin from Saturated Atmosphere*

Time	Data Low	Calculated Intake	Data High
6.5 hrs.	15.27	18.08	
16.5 hrs.		27.61	27.62
24 hrs.		33.53	34.28
48 hrs.		48.16	48.23
72 hrs.	58.43	58.89	
96 hrs.		67.37	67.75
121 hrs.		74.64	74.76
144 hrs.	79.54	80.35	
192 hrs.		90.17	90.20
		Break in curve	
240 hrs.		98.17	98.52
288 hrs.		105.22	106.17
336 hrs.	110.97	111.54	
432 hrs.		122.52	122.54
528 hrs.	131.81	131.82	
624 hrs.	139.50	139.93	
720 hrs.		147.08	147.42
840 hrs.		154.98	155.13
960 hrs.		161.99	163.73
1,128 hrs.	170.59	170.62	

The actual rate of intake has been measured at several points by measuring the tangents to the calculated curve, and we give the rate of intake in grams per minute.

When y = 25 percent, velocity of intake is .015101.
When y = 30 percent, velocity of intake is .013348.
When y = 35 percent, velocity of intake is .011672.
At the break, y = 93 percent, velocity of intake is .002808.

The rate at the time the break occurred was approximately one fifth of the rate when 25 percent had been absorbed. The steady fall in the rate of intake should be noted. These rates are very much lower, of course, than when gelatin is immersed in water, and must depend partly at least on the surface-volume relation of the particular pieces of gelatin used. One would expect a thin, flat piece of gelatin to absorb more rapidly than an equal mass in spherical form.

An important difference between the curve of absorption as shown for gelatin in saturated vapor and that obtained for seeds in water should be noted. The absorption curve for Xanthium seeds shows a break at about 35 percent due to approaching saturation. The gelatin curve here presented shows no such break due to approaching saturation, but maintains a slowly decreasing rate over long periods of time, with remarkable regularity.

DISCUSSION

The data which are presented indicate that gelatin absorbs much more water from a saturated atmosphere than was found by von Schröder. And an examination of his data leads one to suspect that he did not maintain a saturated atmosphere. It is not so easy to see in table 2, but in table 3, which records the loss of water from saturated gelatin, it is rather easily detected. If the behavior in this case were normal, we should find the loss largest during the first 24 hours, because the difference in moisture equilibrium was greatest at the beginning of the experiment. Each day thereafter should show less and less daily loss because of the closer and closer approach of equilibrium conditions. This, however, is not the case in his data. On the first day the loss is 0.259 g., on the second day 0.078 g., on the third day 0.046 g., on the fourth day 0.035 g., and on the fifth day 0.511 g. Counting average daily loss, there was nearly twice as large average daily loss at the end of five days as at the end of the first four days. And at the end of 14 days the average daily loss was still more than double that at the end of the first four days. From the fifth day on, the loss is always much more rapid than during the second, third, and fourth days. This would make it appear very probable that the gelatin was losing water into an unsaturated atmosphere.

And if the atmosphere is unsaturated in this case, it probably was unsaturated in the experiments, the data of which are recorded in table 2. It is a very difficult matter to produce and maintain conditions of saturation, and there is not much doubt that the frequent opening of the chamber for weighing allowed the atmosphere to fall considerably below the saturation point. There is nothing in von Schröder's discussion to show how he handled his materials. Even in our own work we can not be certain that complete saturation was procured and maintained at all times. But the results would indicate that we came nearer to it than did von Schröder.

We feel that the amount of work done is insufficient to show that colloids do not exhibit the phenomenon to which von Schröder's work called attention. Even though we have shown that gelatin takes up from saturated vapor much more water than was formerly supposed, it may still be true that colloids show a difference in behavior toward water in liquid and in gaseous form. Much of the difference in the actual amount of water taken in must be related to the filling of minute lacunae when immersed in water, and the saturation of the gelatin around the lacunae only, when exposed to vapor. The vapor pressure of a saturated colloid may actually be greater than that of a flat surface of water, and the colloid be able therefore to distil water into the saturated atmosphere which is common to both. Whatever the truth may be in regard to this point, it seems to us unfortunate that a theory of physical chemistry should be based upon a single piece of work which shows somewhat gross irregularities in the data, without any attempt to confirm the original findings.

The experience we have had with gelatin in saturated atmosphere, with conditions controlled as carefully as possible, at least suggests the desirability of a reinvestigation of the relation of the vapor pressure of colloids to that of the vapor pressure of water, before we try to establish or accept theories which attempt to explain this relation. It might be found that there is little to explain.

The University of Kentucky,
Lexington, Ky.

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